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MECHANICS OF SUSPENSIONS OF RIGID UNIAXIAL DUMBBELLS IN AN ANISOTROPIC LIQUID

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The structural-phenomenological approach [1, 2] made it possible to obtain [3-11] the rheological equations of dilute suspensions of rigid uniaxial dumbbells in non-Newtonian isotropic disperse media and to study the influence of the non-Newtonian properties of the disperse media on the rheological behavior of the suspensions.

In this study the structural-phenomenological approach was used to construct the rheological equations of dilute suspensions of rigid uniaxial dumbbells in an anisotropic Ericksen liquid [12, 13]. The rheological behavior of such suspensions in a simple slip flow was studied in the presence of an external electric field.

1. Rheological Model of a Disperse Medium. An anisotropic Ericksen liquid [12, 13] is the simplest phenomenological model of an orientable liquid with an unreformed microstructure. The behavior of the microstructure is described by a unit vector n_i, called a director. It characterizes the orientation of particles of the liquid during flow. The stress in the anisotropic Ericksen liquid is a function of the deformation rate tensor and the director:

$$\sigma_{ij} + p\delta_{ij} = 2\mu\gamma_{ij} + \mu_1 n_{ij} + \mu_2\gamma_{lm}n_{lmij} + 2\mu_3(\gamma_{jl}n_{li} + \gamma_{il}n_{lj}). \tag{1.1}$$

In the rheological equation (1.1) p is the isotropic pressure; δ_{ij} is a unit vector; $n_{ij} = n_i n_j$; $n_{\ell mij} = n_\ell n_m n_i n_j$; μ, μ_1, μ_2, μ_3 are phenomenological constants; $\gamma_{ik} = (1/2)(v_{i,k} + v_{k,j})$; $v_{i,k}$ is the derivative of the velocity vector ν_i in the direction of the coordinate of the k axis.

The orientation of the director n_i is determined by the flow and generally depends on the liquid velocity gradient. The defining equation for the director, on the assumption that the inertia of the elements of the microstructure of the anisotropic liquid can be neglected, in the linear approximation in the velocity gradient has the form

$$\frac{Dn_i}{Dt} = \lambda(\gamma_{il}n_l - \gamma_{lm}n_{lmi}), \tag{1.2}$$

where $Dn_i/Dt \equiv \dot{n}_i - \omega_{i\ell}n_\ell$ is the Youman derivative with respect to time; the dot above n_i denotes an individual derivative with respect to time; $\omega_{i\ell} \equiv (1/2)(v_{i\ell} - v_{\ell i})$ is the vorticity tensor; λ is a phenomenological constant; and $n_{\ell mi} = n_\ell n_m n_i$.

According to (1.2), the orientation of the director n_i depends essentially on the dimensionless constant λ. For $|\lambda| < 1$ the director periodically changes in time [12] and its orientation depends on the flow velocity gradients. Equations (1.1) and (1.2) in this particular case were used in [1] to construct a structural-phenomenological theory of the stressed state in a dilute suspension of rigid ellipsoids of rotation with a Newtonian dispersion medium.

In this study we consider anisotropic liquids for which $|\lambda| \geq 1$ and the stress in the state of rest coincides with the isotropic hydrostatic pressure, i.e., liquids for which $\mu_1 = 0$.

The director n_i for $|\lambda| \geq 1$ in steady-state flows assumes a stationary orientation, which is independent of the velocity gradients. Equation (1.2) in simple slip flow

$$v_x = 0,\ v_y = Kx,\ v_z = 0 \quad (K\text{—const}) \tag{1.3}$$

has the stationary solution

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$$\mathrm{ctg}^2\beta = \frac{\lambda - 1}{\lambda + 1},\ \psi = \frac{\pi}{2}, \tag{1.4}$$

which does not depend on the slip velocity K. Here β, ψ are angles that specify the position of the director in the laboratory frame Oxyz ($n_x = \cos\beta \sin\psi$, $n_y = \sin\beta \sin\psi$, $n_2 = \cos\psi$); β is the angle between the Ox axis and the projection of the vector n_ℓ onto the Oxy plane; ψ is the angle between the Oz axis and the vector n_i. The orientation of the director depends on the value of the constant λ. The angle β in (1.4) varies between the limits $\pi/4 \le \beta\ \pi/2$ for $\lambda \ge 1$ and $-\pi/4 \le \beta \le 0$ for $\lambda \le -1$.

The orientation of the director changes with the geometry of the flow. In a uniaxial tensile flow ($v_x = -(q/2)x$, $v_y = -(q/2)y$, $v_z = qz$) the director is oriented collinearly to the Oz for $\lambda \ge 1$ and parallel to the Oxy plane for $\lambda \le -1$. The steady-state orientation of the director does not depend on the strain rate q.

According to [14], the rheological equation (1.1) can be written as a generalized Newton's law

$$\sigma_{ij} + p\delta_{ij} = \eta_{ijkl}\gamma_{kl} \tag{1.5}$$

with a viscosity tensor of the form

$$\eta_{ijkl} = 2\mu I_{ijkl} + \mu_2 n_{ijkl} + 4\mu_3 n_{(i}\delta_{j)(k}n_{l)}, \tag{1.6}$$

where $4n_{(i}\delta_{j)(k}n_{\ell)} = n_i\delta_{jk}n_\ell + n_j\delta_{ik}n_\ell + n_i\delta_{j\ell}n_k + n_j\delta_{ij}n_k$; symmetrization is carried out over the indices in parentheses; and $I_{ijk\ell} = (\delta_{ik}\delta_{j\ell} + \delta_{i\ell}\delta_{jk})/2$ is a unit tensor of rank four.

According to (1.6), the effective viscosity of anisotropic liquids (1.1), (1.2) for $\mu_1 = 0$ and $|\lambda| \ge 1$ in steady-state flows does not depend on the flow velocity gradients. In contrast to the case with Newtonian liquids, however, it does depend on the orientation of the director n_i. This enables us to speak of the anisotropy of the liquid (1.1), (1.2) relative to the direction of the director n_i, which appears when it flows. With allowance for this circumstance in [14] we introduced the longitudinal $\eta_\|$ and transverse $\eta_\perp$ (relative to n_i) viscosity of the liquid (1.1), (1.2). These viscosity coefficient have a particular (basic) importance: the viscosity relative to other directions is expressed in terms of them.

The anisotropy of the viscosity in the liquid (1.1), (1.2) leads to anisotropy of its other characteristics. The existence of the basis viscosities $\eta_\|$ and $\eta_\perp$ determines the existence of different coefficients of translational friction $\zeta_\|$ and transverse $\zeta_\perp$ of a spherical particle during motion in the liquid (1.1), (1.2) along and transverse to the director. According to [15], the drag acting on a spherical particle moving with velocity U_k in an anisotropic liquid (1.1), (1.2) can be written as $-\xi_{ik}U_k$, where

$$\xi_{ik} = \zeta_\perp\delta_{ik} + (\zeta_{||} - \zeta_\perp)n_{ik} \tag{1.7}$$

is the tensor of translational friction of a spherical particle in an anisotropic liquid.

The basic viscosities $\eta_\|$ and $\eta_\perp$ can be given in terms of the rheological constants μ, μ_2, μ_3. For this purpose it is necessary to write the rheological equation for the stresses of the liquid (1.1), (1.2), according to [14], in the form

$$\sigma_{ij} + p\delta_{ij} = 2\eta_\perp\gamma_{ij} + 2(\eta_\perp - \eta_{||})\gamma^*_{ij}$$
$$(\gamma^*_{ij} = 2n_{ijlm}\gamma_{lm} - n_{il}\gamma_{lj} - n_{jl}\gamma_{li})$$

and compare it with (1.1). When $\mu_2 = -4\mu_3$ [14] is taken into account this gives us $\eta_\perp = \mu$, $\eta_\| = \mu + \mu_3$.

Description of the behavior of the microstructure of a liquid medium by means of a director found application in the continuum theory of liquid crystals [16]. The defining equations obtained in [17] and used to describe the dynamic properties of nematic liquid crystals are similar to Eqs. (1.1), (1.2).

2. Structural Theory. The model of a uniaxial dumbbell is used as hydrodynamic model of disperse particles having axial and central symmetry as well as suspensions with an isotropic dispersion medium [2-11]: it is a system of two point centers of the hydrodynamic interaction of the model with the environment, the centers being joined by a rigid link (axis) of length L.

The point center of the dumbbell interacts with the dispersion medium as a spherical particle, i.e., as it flows with velocity u_i past the point center of the dumbbell the anisotropic medium acts on that point with a force given by $\xi_{ij}u_j$ (ξ_{ij} is the tensor of translational friction (1.7)).

It is assumed that on the one hand suspended particles modeled by the dumbbells are of a size such that the anisotropic dispersion medium interacts with them as with hydrodynamic bodies but on the other hand the disperse particles should be sufficiently small so that within the neighborhood of each the velocity v_i of the dispersion medium should be a uniform function of the coordinates, $v_i = \omega_{ik}r_k + \gamma_{ik}r_k$ (r_k is the radius-vector specifying the position of the point in the neighborhood in the laboratory frame $Oxyz$, whose origin coincides with the midpoint of the dumbbell axis). It is also assumed that the disperse particles have zero buoyancy.

With these assumptions, the hydrodynamic forces f_i that the anisotropic dispersion medium exerts on the center of hydrodynamic interaction of the dumbbells are given by $f_i = \xi_{ij}(v_{jk}R_k - \dot{R}_j - V_{0j})$ where v_{0j} is the velocity of migration of the particle center relative to the dispersion medium; R_k is the radius-vector of the point center of the dumbbell resistance, assuming the value $(L/2)\nu_k$ for the other point center; and ν_k is the unit vector characterizing the orientation of the dumbbell disperse particle in the given $Oxyz$ coordinate system. The main vector of the hydrodynamic forces F_i acting on the dumbbell then has the form

$$F_i = -2\xi_{ij}v_{0j}. \tag{2.1}$$

The defining equation for the vector ν_i characterizing the orientation of the dumbbell particle is obtained in much the same way as in [2] by vector multiplication of the equation of rotational motion of the disperse particle $\dot{\mathscr{L}}_i = M_i$ by the vector ν_i:

$$\begin{aligned} I(\ddot{\nu}_i + \dot{\nu}_k\dot{\nu}_k\nu_i) = \frac{1}{2}L^2(\xi_{ij}\gamma_{jk}\nu_k \\ - \xi_{jk}\gamma_{kl}\nu_j\nu_l\nu_i - \xi_{ij}N_j + \xi_{jk}N_k\nu_j\nu_i). \end{aligned} \tag{2.2}$$

Here $\mathscr{L}_i = I[\nu\times\dot{\nu}]$ is the angular momentum of the particle; I is the moment of inertia of the dumbbell particle relative to the axis passing through the center of the dumbbell axis and perpendicular to it; $M_i = (1/2)L^2\varepsilon_{ijk}\nu_j(\xi_{k\ell}v_{\ell,s}\nu_s - \xi_{k\ell}\dot{\nu}_\ell)$ is the main moment of hydrodynamic forces relative to the midpoint of the dumbbell axis; ε_{ijk} is the Levi-Civita tensor; and $N_i = \dot{\nu}_i - \omega_{ik}\nu_k$.

For $\zeta_\parallel = \zeta_\perp = \zeta$ Eq. (2.2) coincides with the defining equation for a vector ν_i obtained in [2] for suspensions with an isotropic dispersion medium.

The inertial forces and the moment of the inertial forces of disperse particles in suspensions are very small and are usually ignored in the rheology of suspensions. In this case the equation of translational motion for a disperse particle is $F_i = 0$, from which, according to (2.1), it follows that $\nu_{0j} = 0$, i.e., the disperse particles do not migrate relative to the dispersion medium. Equation (2.2) becomes

$$\begin{aligned} (\xi_{im} - \xi_{km}\nu_k\nu_i + \zeta_\perp\nu_m\nu_i)(\dot{\nu}_m - v_{mk}\nu_k) \\ + \zeta_\perp\gamma_{km}\nu_k\nu_m\nu_i = 0. \end{aligned} \tag{2.3}$$

In much the same way as in [2] within the framework of the structural theory we find an expression for the rate of dissipation of mechanical energy per unit volume of the suspension under consideration,

$$W = \tilde{W} + N_0(L^2/2)[\xi_{ij}\langle N_iN_j\rangle - 2\xi_{ij}\gamma_{jk}\langle N_i\nu_k\rangle + \xi_{ij}\gamma_{ik}\gamma_{jl}\langle \nu_l\nu_k\rangle], \tag{2.4}$$

where $\tilde{W}$ is the rate of energy dissipation per unit volume of dispersion medium in the absence of disperse particles; N_0 is the number of disperse particles per unit volume of suspension; the angular brackets denote averaging, which can be done with the aid of the distribution function of the angular positions of the vector ν_i, which is a solution of the equation

$$\frac{\partial F}{\partial t} + \frac{\partial}{\partial \nu_i}(F\dot{\nu}_i) = 0. \tag{2.5}$$

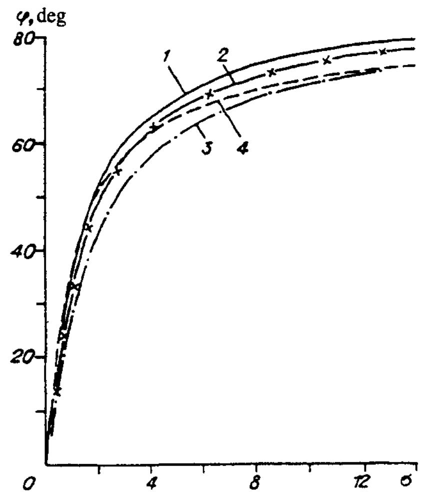

Fig. 1

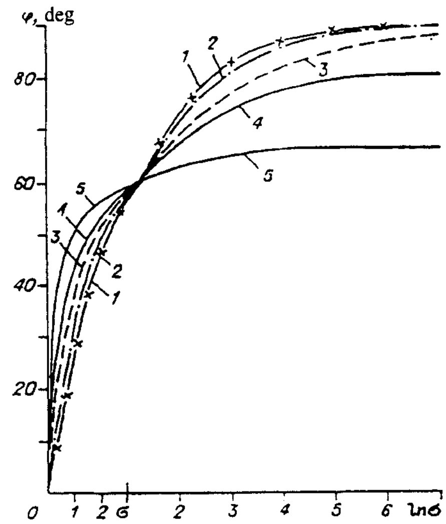

Fig. 2

3. Structural – Phenomenological Theory. According to (2.4), the stress tensor Σ_{ij} in the suspension considered should be determined by relations of the type $\Sigma_{ij} = \sigma_{ij} + N_0 < t_{ij} >$, $t_{ij} = t_{ij}(\gamma_{km}, \xi_{\ell n}, \nu_s, N_p)$ (σ_{ij} is the stress tensor in the dispersion medium in the absence of suspended particles, $N_0 < t_{ij} >$ is the stress caused by the presence of N_0 suspended particles per unit volume of suspension). The tensor t_{ij} should be a polynomial function of the matrix of its arguments, which is linear in $\xi_{km}\gamma_{m\ell}$ and $\xi_{km}N_m$. By virtue of the symmetry of the dumbbell particle about its center that function, furthermore, should be invariant under reversal of the direction of ν_s. The phenomenological equation for t_{ij} is obtained in a way similar to that in [13]:

$$t_{ij} = a_0\delta_{ij} + a_1\nu_i\nu_j + a_2\xi_{lk}\gamma_{lm}\nu_k\nu_m\nu_i\nu_j + a_3\xi_{in}\gamma_{nj} + a_4\xi_{jn}\gamma_{ni} + a_5\xi_{ln}\gamma_{ln}\nu_i\nu_j$$
$$+ a_6\xi_{ln}\gamma_{jn}\nu_l\nu_i + a_7\xi_{in}\gamma_{nk}\nu_k\nu_j + a_8\xi_{jn}\gamma_{nk}\nu_k\nu_i + a_9\nu_i\xi_{jk}N_k + a_{10}\nu_j\xi_{ik}N_k. \quad (3.1)$$

The phenomenological constants a_i ($i = \overline{1,10}$) in (3.1) are found by comparing the dissipation rate of mechanical energy per unit volume of suspension (2.4) determined in the structural theory, and the rate determined as in [2] from the formula $W = W + N_0 < t_{ij}\gamma_{ij} > - N_0 < N_i g_i >$ within the framework of the phenomenological approach, where g_i is the right side of (2.2). Taking $< t_{ij}\nu_j > - < t_{ji}\nu_j > = < g_i >$ into account, we obtain $a_1 = a_2 = a_3 = a_4 = a_5 = a_6 = a_8 = a_9 = 0$, $a_7 = L^2/2$, $a_{10} = -L^2/2$, which allows us finally to write the defining equation for the stress tensor Σ_{ij}:

$$\Sigma_{ij} = \sigma_{ij} + (1/2)N_0L^2(\xi_{il}\gamma_{mk}\langle\nu_k\nu_j\rangle - \xi_{jk}\langle\nu_iN_k\rangle). \quad (3.2)$$

If the disperse particles, modeled by dumbbells, have a constant dipole moment $p_i = q\nu_i$, then in an external uniform electric field E_i such particles are acted upon by a couple with moment $M_i^e = q\varepsilon_{i\ell m}\nu_\ell E_m$, tending to turn the particle in the direction of the electric vector E_i.

If the disperse particles are sufficiently small, we must take into account their rotational Brownian motion caused by the effective moment of the forces

$$M_i^B = -T\varepsilon_{ilm}\nu_l\partial\ln F/\partial\nu_m$$

(T is the temperature in units of energy). When the moment of the inertial forces of the disperse particles is not taken into consideration the defining equation for the vector ν_i has the form

$$\frac{L^2}{2}(\xi_{im} - \xi_{km}\nu_k\nu_i + \zeta_\perp\nu_i\nu_m)(\dot\nu_m - \upsilon_{mk}\nu_k)$$
$$+ \frac{L^2}{2}\zeta_\perp\gamma_{km}\nu_k\nu_m\nu_i - q(E_i - E_k\nu_k\nu_i) + \bar{T}\left(\frac{\partial\ln F}{\partial\nu_i} - \nu_i\nu_k\frac{\partial\ln F}{\partial\nu_k}\right) = 0. \quad (3.3)$$

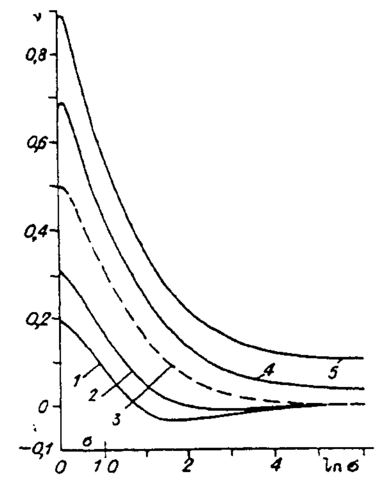

Fig. 3

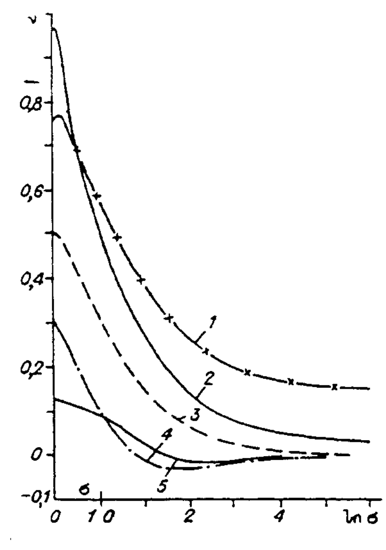

Fig. 4

Equations (1.1), (1,2), (2.3) (or (3.3)), (2.5), (3.2) constitute a closed system of equations defining the stressed state in a dilute suspension of dumbbell particles in an anisotropic dispersion medium.

4. Steady-State Flow of Simple Shear. An example of the use of the rheological equations obtained we study the effect of the anisotropy of a dispersion medium on the rheological behavior of a dilute suspension of dipole dumbbells in an anisotropic liquid (1.1), (1.2) for $|\lambda| \geq 1$, $\mu_1 = 0$ in simple shear flow (1.3) in an electric field $E_x = E$, $E_y = 0$, $E_z = 0$ (E is constant). The solution (1.4) of Eq. (1.2) shows that the electrically neutral director n_i of an anisotropic dispersion medium (1.1), (1.2) is oriented in the steady state in planes perpendicular to the Oz axis at an angle β to the Oxz coordinate plane; β is determined by (1.4).

It follows from (3.3) that dipole disperse dumbbell particles also assume a steady-state orientation in both an isotropic ($\Delta \approx 1$) dispersion medium in a plane perpendicular to the Oz axis at an angle φ to the Oxz plane, which is determined by the equation

$$\cos^2\varphi - (\Delta - 1)\cos\varphi\sin\beta\sin(\varphi - \beta) - \frac{1}{\sigma}\sin\varphi = 0, \tag{4.1}$$

where $\sigma = \frac{K\gamma}{qE}$; $\gamma = \frac{\zeta_\perp L^2}{2}$; $\Delta = \frac{\zeta_\parallel}{\zeta_\perp}$.

Figures 1 and 2 show graphs of $\varphi = \varphi(\sigma)$. Curves 1-3 of Fig. 1 correspond to $\beta = 45$, 60, and 75° for $\Delta = 0.5$ and curves 1-5 of Fig. 2 correspond to $\Delta = 0.2$, 0.5, 1, 1.5, and 5.0 for $\beta = 60°$. Curve 4 (Fig. 1) and 3 (Fig. 2) corresponds to an isotropic dispersion medium ($\Delta = 1$).

We found that as σ increases the hovering angle of the dumbbell particle increases from $\varphi_0 = 0$ for $\sigma = 0$ to φ_∞, whose value depends on the anisotropy of the medium: $\varphi_\infty = 90°$ for $\Delta \leq 1$ and $\varphi_\infty = \mathrm{arctg}\{(1 + \Delta\mathrm{tg}^2\beta)/[(\Delta - 1)\mathrm{tg}\beta]\}$ for $\Delta > 1$. The hovering angle of dumbbell particles in an anisotropic medium in which $\Delta < 1$ is smaller than in an isotropic medium ($\Delta = 1$) when $\varphi < \beta$, and larger when $\varphi > \beta$ (Fig. 1); the reverse is true in an anisotropic medium in which $\Delta > 1$ (Fig. 2).

With the disperse particles in a steady-state orientation the distribution of their angular positions, which generally is found from the solution of Eq. (2.5), is transformed into a Dirac delta function concentrated in the hovering angle φ. The effective viscosity of the suspension, determined from (3.2), therefore, becomes

$$\mu_a = \mu_E + \frac{1}{2}N_0\gamma[\cos^2\varphi + (\Delta - 1)\cos\varphi\sin\beta\sin(\varphi + \beta)], \tag{4.2}$$

where $\mu_a \equiv (\Sigma_{xy} + \Sigma_{yx})/2K$; $\mu_E = \mu + \mu_2(\lambda^2 - 1)/4\lambda^2 + \mu_3$ is the effective viscosity of the anisotropic dispersion medium (1.1), (1.2) for $|\lambda| \geq 1$, $\mu_1 = 0$ in a simple shear flow (1.3).

According to (4.2), the anisotropic properties of the dispersion medium for $\Delta > 1$ increase the increment $\nu = (\mu_a - \mu_E)/N_0\gamma$ of the effective viscosity μ_a of the suspension, and for $\Delta < 1$ decreases the increment, in comparison with an isotropic ($\Delta = 1$) dispersion medium (Figs. 3, 4).

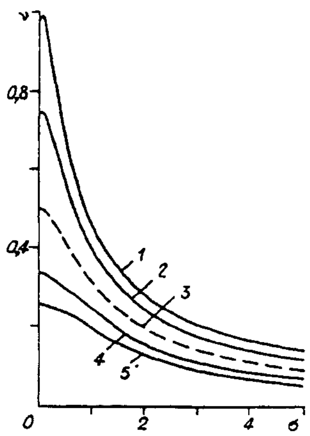

Fig. 5.

Figures 3 and 4 show the graph of $\nu = \nu(\sigma)$. Curves 1-5 of Fig. 3 correspond to $\Delta = 0.2, 0.5, 1$ (isotropic dispersion medium); 1.5, 2 for $\beta = 60°$; curves 1, 2, Fig. 4 – $\beta = 45, 74°$ for $\Delta = 2$, curve 3 – $\Delta = 1$ (isotropic dispersion medium), curves 4, 5 – $\beta = 45, 75°$ for $\Delta = 0.2$.

For $\Delta > 1$ and small σ (curves 4, 5 of Fig. 3 and curves 1, 2 of Fig. 4) and for $\Delta < 1$ and large σ (curves 1, 2 of Fig. 3 and curves 4, 5 of Fig. 4) we note segments where the viscosity increments grow as σ increases; those segments are not intrinsic to dilute suspensions of rigid particles with an isotropic dispersion medium ($\Delta = 1$).

When the suspended particles reach their limiting orientation φ_∞ the viscosity increment reaches its limiting value ν_∞ which depends on the rheological parameters of the dispersion medium. For $\Delta > 1$ the limiting values of the increment ν_∞ are nonzero (curves 4, 5 of Fig. 3 and curves 1, 2 of Fig. 4). At the same time for $\Delta = 1$ (isotropic dispersion medium) and $\Delta < 1$, as in [2-11], $\nu_\infty = 0$ (curves 1-3 of Fig. 3 and curves 3-5 of Fig. 4). This means that the use of uniaxial dumbbells as a hydrodynamic model of disperse particles, making it possible to predict the existence of a limiting value of the increment of the effective viscosity ν_∞ as $\sigma \to \infty$ is smaller than the increment ν_0 as $\sigma \to 0$, results in an underestimation of its values. This drawback of the theory can be eliminated only by using a hydrodynamic model having volume (ellipsoid of rotation) or transverse dimensions (triaxial dumbbell).

Formulas (4.1), (4.2) were used to determine $\nu = \nu(\sigma)$ in a dilute suspension of dumbbell dipole particles in *N*-(*n*-methoxybenzilidene)-*n*-butylaniline (MBBA). It is known [16] that MBBA, being a nematic liquid crystal, at 22°C is an anisotropic liquid with $\lambda = 1.04$, $\beta = 82°$. Since no experimental data on $\zeta_\parallel$ and $\zeta_\perp$ are available for MBBA, in Fig. 5 we show $\nu = \nu(\sigma)$ for various values of Δ. Curves 1-5 of Fig. 5 correspond to $\Delta = 2.0, 1.5, 1$ (isotropic dispersion medium), 0.66, and 0.5.

The results obtained in this study can be used to construct a theory of dynamic behavior of impurities in liquid crystals as well as the influence of impurities on the rheological behavior of liquid crystals.

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